

# PATENT ABSTRACTS OF JAPAN

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## (54) ROOM TEMPERATURE CURING COMPOSITION AND ITS PRODUCTION

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To produce a room temperature curing composition, comprising a specific polymer and a specified low-molecular polymer, having a sufficient low viscosity without deteriorating the flexibility of a cured product even when using an inorganic filler and useful as a sealing material, etc.

**SOLUTION:** This room temperature curing composition comprises (A) 100 pts.wt. polymer having 1-1.5 hydrolyzable silicon groups on the average in one molecule and 8000-50000 molecular weight and (B) 1-200 pts.wt. low-molecular polymer having 0.5-1.5 hydrolyzable silicon groups on the average in one molecule and 300-8000 molecular weight. Furthermore, both the main chains of the components (A) and (B) are essentially polyethers and the hydrolyzable silicon groups of the components (A) and (B) are represented by the formula  $R_2-SiXaR_{13-a}$  [R1 is a 1-20C (substituted)monovalent organic group; R2 is a bivalent organic group; X is OH or a hydrolyzable group; (a) is 1-3]. The composition preferably contains further (C) a plasticizer, especially without containing a low-molecular one or preferably does not contain substantially the component (C).

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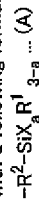
## CLAIMS

[Claim(s)]

[Claim 1]As opposed to polymer with an or more 8000 molecular weight [ or less 50000 ] (I) and polymer (I)100 weight section which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups, A room-temperature-curing nature constituent containing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II)1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups.

[Claim 2]A room-temperature-curing nature constituent of Claim 1 whose both main chain of polymer (I) and main chain of low molecule polymer (II) are polyether intrinsically.

[Claim 3]Claim 1 or 2 room-temperature-curing nature constituents by which both a hydrolytic silicon group of polymer (I) and a hydrolytic silicon group of low molecule polymer (II) are expressed with a following formula (A).



$R^1$  is a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted among a formula (A).  $R^2$  is a divalent organic group, X is a hydroxyl group or a hydrolytic basis, and a is an integer of 1-3.

[Claim 4]Claim 1, a room-temperature-curing nature constituent of 2 or 3 in which a room-temperature-curing nature constituent does not contain a plasticizer substantially.

[Claim 5]Claim 1, a room-temperature-curing nature constituent of 2 or 3 which a room-temperature-curing nature constituent contains a plasticizer further, and do not contain a low molecule plasticizer as the plasticizer.

[Claim 6]As opposed to polymer (I)100 with an or more 8000 molecular weight [ or less 50000 ] weight section which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups, A manufacturing method of a room-temperature-curing nature constituent mixing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II) 1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups.

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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the room-temperature-curing nature constituent hardened under hygroscopic-surface-moisture existence.

[0002]

[Description of the Prior Art] The method of making harden various kinds of polymers which have an end hydrolytic silicon group, and using it for a sealing material, adhesives, etc. is known well, and is a useful method industrially.

[0003] The polymer especially whose main chain is polyether among such polymers is liquefied at a room temperature, and when a hardened material holds pliability also at low temperature comparatively and uses for a sealing material, adhesives, etc., it is provided with the desirable characteristic.

[0004] As a polymer of such hygroscopic-surface-moisture hardenability, the polymer of the hygroscopic-surface-moisture hardenability which has a hydrolytic silicon group is mentioned to the end indicated to JP.3-72527.A, JP.3-47825.A, etc. In the polymer which has a hydrolytic basis at such the end, although the pliability of a hardened material increases, the viscosity of resin becomes high and workability gets remarkably bad, so that the molecular weight is generally large.

[0005] If the molecular weight of such a polymer is small, viscosity will become low, but a hardened material becomes a thing inferior to pliability. In order to make a polymer into hypoviscosity, maintaining the pliability of a hardened material until now, various kinds of plasticizers have been used.

[0006] As such a plasticizer, aromatic carboxylic acid ester, aliphatic-carboxylic-acid ester species, glycol ester, phosphoric ester, an epoxy plasticizer, a chlorinated paraffin, etc. are used. However, since these plasticizers are translatable, when it is used for a sealing material etc., there is a fault which has the surface contamination after contamination and paint of the ceiling part circumference and an adverse effect to an adhesive property.

[0007] The hardenability constituent which adds the very low reactant plasticizer of translatability is proposed by JP.5-59267.A, without reducing the pliability of a hardened material to the polymer of the hygroscopic-surface-moisture hardenability which has a hydrolytic silicon group in order to cancel such a fault. However, in the use to a sealing material etc., pliability is higher, and when it paints on the surface in the good characteristic of elongation, the hardenability constituent in which surface stain resistance has been improved is called for. Although various kinds of inorganic bulking agents are usually used in combination called a actual sealing material and adhesives, a polymer's own pliability is much more required for use of a bulking agent in order to harden the physical properties of a hardened material.

[0008]

[Problem(s) to be Solved by the Invention] Then, even if the viscosity of the hardenability constituent could be reduced and it used the inorganic bulking agent, the hardened material with a flexibly good extension characteristic was given, and moreover, translatability was dramatically low, and as a result of examining the additive agent which does not pollute a surface coat, it resulted in this invention.

[0009]

[Means for Solving the Problem] Namely, polymer with an or more 8000 molecular weight [ or less 50000 ] (I) and polymer (II) 100 weight section which averages this invention in one molecule and has

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1.5 or less one or more piece hydrolytic silicon groups is received, A room-temperature-curing nature constituent containing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II) 1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups, And polymer (I) 100 with an or more 8000 molecular weight [ or less 50000 ] weight section which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups is received, a manufacturing method of a room-temperature-curing nature constituent mixing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II) 1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups -- it comes out.

[0010]

[Embodiment of the Invention] As for both polymer (I)s and low molecule polymer (II)s that are used by this invention, consisting of polyether intrinsically is [ the main chain of a molecule ] preferred.

[0011] As for such a polymer, what is obtained by introducing a hydrolytic silicon group by the suitable method for a polyether containing hydroxyl group is preferred.

[0012] Such a polymer is proposed by JP.3-47825.A, JP.3-72527.A, JP.3-79627.A, JP.46-30711.B, JP.45-36319.B, JP.46-17553.B, etc., for example.

[0013] A polyether containing hydroxyl group is obtained by polymerizing monoepoxide, such as bottom alkylene oxide of existence of an initiator and a catalyst.

[0014] The compound which has 1-10 active hydrogen as an initiator is preferred. As a compound which has 2-10 active hydrogen, a polyhydroxy compound is preferred, and the polyhydroxy compound which has 2-4 hydroxyl groups especially is preferred 2-8 pieces. Specifically Ethylene glycol, a diethylene glycol, propylene glycol, Dipropylene glycol, neopentyl glycol, 1,4-butanediol, There is polyol of low molecular weight from the object produced by making monoepoxide react to 1,6-hexanediol, glycerin, trimethylolpropane, pentaerythritol, diglycerol, a shock sirfain, and these. One-sort single use or two or more sorts of concomitant use may be sufficient as these.

[0015] As a compound which has one active hydrogen, an unsaturation group content mono- hydroxy compound, for example like allyl alcohol is preferred. When manufacturing especially low molecule polymer (II), the aliphatic series of the carbon numbers 1-20, alicyclic fellows and an aromatic monooar, a thiol, secondary amine, carboxylic acid, etc. can be used.

[0016] As monoepoxide, there are propylene oxide, butylene oxide, ethylene oxide, allyl glycidyl ether, etc. Especially propylene oxide is preferred.

[0017] As a catalyst, catalysts, such as an alkaline metal catalyst, a composite metal cyanide complex catalyst, and metalloporphyrin, are mentioned.

[0018] Especially desirable polyethers containing hydroxyl group are polyoxypropylene diol, polyoxypropylene triol, polyoxypropylene tetraol, and polyoxypropylene hexaol. When using for following (1) or the method of (4), polyether of olefin ends, such as polyoxypropylene glycol monoallyl ether, can also be used.

[0019] The hydrolytic silicon group should just be a silicon group with which hydrolysis and crosslinking reaction occur with hygroscopic surface moisture. The silicon content group which has the hydrolytic basis coupled directly with the silicon atom can be used. For example, the basis expressed with a formula (A) is preferred.

[0020]  $R^2-SiX_3-R^1 \dots (A)$

[0021]  $R^1$  is a univalent organic group the substitution of the carbon numbers 1-20, or unsubstituted among a formula (A),  $R^2$  is a divalent organic group, X is a hydroxyl group or a hydrolytic basis, and a is an integer of 1-3.

[0022] As  $R^1$  in a formula (A), a with a carbon number of eight or less alkyl group, a phenyl group, and a fluoro alkyl group are preferred, and a methyl group, an ethyl group, a propyl group, a butyl group, a hexyl group, a cyclohexyl group, especially a phenyl group, etc. are preferred.

[0023] X is a hydroxyl group or a hydrolytic basis, and there are a halogen atom, an alkoxy group, an acyloxy group, an amide group, an amino group, an aminoxy group, a KETOKISHI mate group, and a hydride group as a hydrolytic basis, for example. As for especially the carbon number of the hydrolytic basis which has a carbon atom among these, four or less are [ six or less ] preferred. As desirable X, a with a carbon number of four or less lower alkoxy group especially a methoxy group, an

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ethoxy basis, a propoxy group, etc. can be illustrated, a is an integer of 1-3 and 2 or 3 is preferred. [0024]Next, the manufacturing method of polymer (I) and (II) is explained. These polymers introduce a hydrolytic silicon group into the end of a polyether containing hydroxyl group by a method like following the (1) - (4), and are manufactured.

[0025](1) A method to which the silicon hydride compound expressed with what introduced the unsaturation group into the end of hydroxyl group end polyether, and a formula (B) is made to react under existence of a catalyst. However, R<sup>1</sup> in formula (B), X, and a are the same also in a formula (A).

[0026]HSiX<sub>3</sub>R<sup>1</sup><sub>3-a</sub>... (B)

[0027]Here, an unsaturation group is introduced or more into one of the end of hydroxyl group end polyether with what introduced the unsaturation group into the end of hydroxyl group end polyether. After setting terminal hydroxyl groups OH of hydroxyl group end polyether to OM (M is an alkaline metal) as this method, There is the method of making the compound which has a functional group which can react to the method or unsaturation group, and hydroxyl group which are made to react to unsaturation group content halogenated hydrocarbon, such as an allyl chloride, react to hydroxyl group end polyether, and combining by the ester bond, a urethane bond, carbonate combination, etc. [0028]When polymerizing monoepoxide in manufacture of hydroxyl group end polyether, it is obtained also by using a terminal unsaturated group content mono- hydroxy compound as the method of introducing an unsaturation group into a side chain, or an initiator by carrying out copolymerization of the unsaturation group content monoepoxide, such as allyl glycidyl ether.

[0029](2) How to make the compound which has a hydrolytic silicon group expressed with an isocyanate group and a formula (A) react to hydroxyl group end polyether.

[0030](3) A method to which W basis of the silicon compound expressed with a formula (C) to this isocyanate group is made to react after making polyisocyanate compounds, such as tolylene diisocyanate, react to hydroxyl group end polyether and considering it as an isocyanate group end.

[0031]R<sup>1</sup><sub>3-a</sub>-SiX<sub>a</sub>-R<sup>3</sup>W... (C)

[0032]However, also in a formula (A), R<sup>1</sup> in formula (C), X, and a are the same, R<sup>3</sup> is a divalent organic group, and W is the active hydrogen containing group chosen from the hydroxyl group, the carboxyl group, the sulphydryl group, and the amino group (the 1st class or the 2nd class).

[0033](4) A method to which the unsaturation group of what introduced the unsaturation group into the end of hydroxyl group end polyether, and the sulphydryl group of a silicon compound expressed by the formula (C) whose W is a sulphydryl group are made to react.

[0034]Especially in manufacture of low molecule polymer (II), the molecular terminal group besides hydrolytic silicon Motomochi may be an inertness organic group. For example, it may manufacture by changing into an inertness organic group by the method of the terminal hydroxyl groups making a terminal unsaturated group react to a hydrolytic silicon group with a benzoyl chloride etc. by using as a raw material the polyether monooar which manufactured the terminal unsaturated group content mono- hydroxy compound as an initiator.

[0035]Polymer (I) in this invention is a with an or more 8000 molecular weight [ or less 50000 ] which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups polymer. As for the molecular terminal cardinal number per molecule of polymer (I), 2-8 are preferred, and 2-especially 4 are preferred.

[0036]The molecular weight of polymer (I) is 50000 or less [ 8000 or more ]. When the molecular weight of this organic polymer is lower than 8000, the elongation of a hardened material becomes low, and when a molecular weight exceeds 50000, workability gets remarkably bad for hyperviscosity. As for a molecular weight, it is preferred that it is 10000-40000, and it is preferred that it is especially 10000-30000.

[0037]The molecular weight of polymer (I) and low molecule polymer (II) is computed based on the hydroxyl value conversion value molecular weight of hydroxyl group end polyether which is a raw material.

[0038]Low molecule polymer (II) is a with an or more 300 molecular weight [ less than 8000 ] which averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups low molecule polymer. The hydrolytic silicon cardinal number in low molecule polymer (II) is averaged in a chain, and 1.2 or less [ 0.5 or more ] are preferred. As for the molecular terminal cardinal number per one molecule of low molecule polymer (II)s, 2-8 are preferred, and 2-especially 4 are preferred.

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[0039]The molecular weight of low molecule polymer (II) is less than [ 300 or more ] 8000. When a molecular weight is 8000 or more, the effect as an adhesiveness-reducing agent becomes low. 300-6000 are preferred and 2000-4000 are more preferred.

[0040]Low molecule polymer (II) plays a role of the reactant adhesiveness-reducing agent or plasticizer of non-translatibility.

[0041]In this invention, 200 or less weight section one or more-weight section of low molecule polymer (II)s are used to polymer (I)100 weight section. One to 100 weight section is preferred, and especially one to 80 weight section is preferred.

[0042]It is preferred to mix and use low molecule polymer (II) to polymer (I) in this invention.

[0043]In the constituent of this invention, if necessary to publicly known various curing catalysts, a bulking agent, an additive agent, and a pan, a solvent, a plasticizer, etc. can be included.

[0044]The following compound can be used as a curing catalyst. Metal salt, such as an alkyl titanate, an organic silicon titanate, and bismuth tris-2-ethylhexoate. Acidic compounds, such as phosphoric acid, p-toluenesulfonic acid, and phthalic acid, a butylamine, Aliphatic series monoamines, such as hexylamine, octylamine, decyl amine, and lauryl amine. Aliphatic diamine, such as ethylenediamine and a hexanediamine, diethylenetriamine, Aliphatic polyamine, such as triethylenetetramine and tetraethylenepentamine. Amine compounds, such as aromatic amine, such as heterocyclic amine, such as piperidine and a piperazine, and a meta-phenylenediamine, ethanalamines, triethylamine, and various denaturation amine used as a hardening agent of an epoxy resin.

[0045]Divalent tin and the mixture of the above-mentioned amines of tin dioctylate, JINAFUTEN \*\*\* distearic acid tin, etc.

[0046]Dibutyltin diacetate, dibutyltin dilaurate, dioctyl tin dilaurate, the following carboxylic type organic tin compound, and the mixture of these carboxylic type organic tin compounds and the above-mentioned amines. (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Sn(OOCCH=CHCOOCH<sub>3</sub>)<sub>2</sub>, (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Sn

(OOCCH=CHCOOOC<sub>4</sub>H<sub>9</sub>-n)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn(OOCCH=CHCOOCH<sub>3</sub>)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn

(OOCCH=CHCOOOC<sub>4</sub>H<sub>9</sub>-n)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn(OOCCH=CHCOOOC<sub>8</sub>H<sub>17</sub>-iso)<sub>2</sub>

[0047]The following sulfur-containing mold organic tin compound. (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Sn (SCH<sub>2</sub>COO), (n-

C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn (SCH<sub>2</sub>COO), (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn (SCH<sub>2</sub>CH<sub>2</sub>COO), (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn

(SCH<sub>2</sub>COOCH<sub>2</sub>CH<sub>2</sub>COOCH<sub>2</sub>S), (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Sn(SCH<sub>2</sub>COOOC<sub>8</sub>H<sub>17</sub>-iso)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn

(SCH<sub>2</sub>COOOC<sub>8</sub>H<sub>17</sub>-iso)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn(SCH<sub>2</sub>COOOC<sub>8</sub>H<sub>17</sub>-n)<sub>2</sub>, and (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>SnS.

[0048](n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Organic tin oxide, such as SnO and (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>SnO. And the resultant of these

organic tin oxide and ester compounds, such as ethyl silicate, dimethyl maleate, a diethyl maleate, dioctyl maleate, dimethyl phthalate, diethyl phthalate, and dioctyl phthalate. [0049]Chelate tin compounds, such as the following, and the resultant of these tin compounds and alkoxysilane (however, acac acetylacetonate ligand). (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>Sn(acac)<sub>2</sub>, (n-C<sub>8</sub>H<sub>17</sub>)<sub>2</sub>Sn(acac)<sub>2</sub>, and

(n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>(C<sub>8</sub>H<sub>17</sub>O)Sn (acac).

[0050]The following tin compound. (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>(OH<sub>3</sub>COO)SnOSn(OOCH<sub>3</sub>)(n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>, (n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>(CH<sub>3</sub>O)SnOSn(OCH<sub>3</sub>)(n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>, (CH<sub>3</sub>O)SnOSn(OCH<sub>3</sub>)(n-C<sub>4</sub>H<sub>9</sub>)<sub>2</sub>.

[0051]If it is considered as a bulking agent, the publicly known following bulking agent can be used.

The calcium carbonate which carried out the surface treatment of the surface with fatty acid or a resin acid system organic matter. Colloid calcium carbonate with a mean particle diameter of 1 micrometer or less which furthermore impalpable-powder-sized this, Calcium carbonate, such as precipitated calcium carbonate with a mean particle diameter of 1-3 micrometers manufactured with the sedimentation method, and heavy calcium carbonate with a mean particle diameter of 1-20 micrometers, Fumes silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, and carbon black, Magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, a mill balloon, wood flour, pulp, a cotton chip, mica, and the blacking wash farina --- rubbing --- powder state bulking agents, such as farina, graphite, aluminium impalpable powder, and the Flint powder. Fibrous fillers, such as asbestos,

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glass fiber, a glass filament, carbon fiber, the Kevlar textiles, and a polyethylene fiber.

[0052]As for especially the amount of the bulking agent used, 50 to 250 % of the weight is preferred one to 1000% of the weight to the sum total of polymer (I) and low molecule polymer (II). These bulking agents may be used independently and may be used together two or more sorts.

[0053]A plasticizer may be used, although the room-temperature-curing nature constituent in this invention is fully hypoviscosity in itself and it is preferred not to use a plasticizer substantially.

[0054]As a plasticizer, for example Diethyl phthalate, dibutyl phthalate, Phthalic acid alkyl ester, such as phthalic acid benzyl ester; Diethyl adipate, Glucitol ester species, such as aliphatic-carboxylic-acid alkyl ester; pentaerythritol ester, such as succinic acid diisodecyl, dibutyl sebacate, and butyl oleate; Triethyl phosphate, Phosphoric ester, such as trisecyl phosphate; epoxy plasticizer, chlorinated paraffin, such as epoxidized soybean oil and epoxy stearic acid benzyl, etc. can be independent, or can use it with two or more sorts of mixtures.

[0055]However, as for a low molecule plasticizer, since a low molecule plasticizer has the problem which is this invention of being easy to carry out after [room-temperature-curing nature constituent hardening] bleed out, among such plasticizers, it is preferred not to use it. That is, it is preferred for the room-temperature-curing nature constituent of this invention to contain a plasticizer further, and not to contain a low molecule plasticizer as the plasticizer. The compound itself is low molecular weight, and a low molecule plasticizer refers to the plasticizer which does not have a reactant group. For example, it is phthalic acid alkyl ester.

[0056]In the constituent of this invention, a hydrolytic silicon compound may be arbitrarily added in order to adjust physical properties and hardenability of a hardened material. As such a compound, specifically Tetramethyl silicate, vinyltrimethoxysilane, Although the compound etc. in which methyl trimethoxysilane, dimethyldimethoxysilane, trimethylmethoxysilane, etc. and these methoxy groups were replaced by the ethoxy basis can be illustrated, it is not limited to these.

[0057]As an additive agent, the photoresist compound aiming at adhesion grant agents, such as a thiotropy grant agent, phenol resin, an epoxy resin, etc. and various kinds of silane coupling agents, paints, various kinds of stabilizer, and surface treatment like oligoester acrylate etc. are mentioned. A solvent can also be used in order to prepare viscosity.

[0058]The room-temperature-curing nature constituent of this invention can be used as a sealing material especially an elastic sealing compound, and adhesives.

[0059]

[Example]Although an example explains this invention below, this invention is not limited to these.

Below, a part shows a weight section. As for the example of manufacture of low molecule polymer a-d, and Examples 5-9, working example of this invention and Examples 14-18 of the example of manufacture of polymer A-E and Examples 10-13 are [Examples 1-4] comparative examples.

[0060][Example 1] Propylene oxide was made to react under existence of a zinc hexa cyanocobaltate complex compound catalyst by having used 2-ethylhexanol as the initiator, and the polyoxypropylene monooar was manufactured. Subsequently, after using 95% of terminal hydroxyl groups as an allyloxy group, methyl dimethoxysilane of the equivalent was made to react 80% to an allyl group by having made chloroplatinic acid into the catalyst further, and the polymer a of the molecular weight 5000 [about ] which has a methyl dimethoxy silyl propyl group was compounded. Viscosity was 100cP at 25 \*\*.

[0061][Example 2] The bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst was made to react by having used 2-ethylhexanol as the initiator, and the polyoxypropylene monooar was manufactured. The methyl dimethoxy silyl propylisocyanate of equimolar was made to react to this furthermore, and the polymer b of the molecular weight 4000 [about ] was compounded. Viscosity was 1050cP at 25 \*\*.

[0062][Example 3] Propylene oxide was made to react by having used allyl alcohol as the initiator, and the polyoxypropylene monooar of the molecular weight 2000 was manufactured. Then, it was made to react to a hydroxyl group and the benzoyl chloride of equimolar under triethylamine existence. The reaction mixture was diluted with 5-times the amount hexane, and was rinsed, except for the triethylamine hydrochloride, hexane was distilled off and one end acquired the monoallyl object of the benzoyloxy group. Next, it was made to react to methyl dimethoxysilane of the equivalent 80% to an allyl group by having made chloroplatinic acid into the catalyst, and the polymer c of the molecular weight 2200 which has a methyl dimethoxy silyl propyl group was compounded.

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Viscosity was 700cP at 25 \*\*.

[0063][Example 4] Propylene oxide was made to react by having used butanol as the initiator, and the polyoxypropylene monooar of the molecular weight 6000 was manufactured. After using 93% of terminal hydroxyl groups as an allyloxy group, the dimethylethoxy silane of the equivalent was made to react 95% to an allyl group, and the polymer d of the molecular weight 6000 [about ] which has a dimethylethoxy silylpropyl group was compounded. Viscosity was 2500cP at 25 \*\*.

[0064][Example 5] Ethylene glycol was used as the initiator and the terminal hydroxyl groups of the polyoxypropylene diol produced by making the bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst react was changed into the allyloxy group. Methyl dimethoxysilane was made to react by having made chloroplatinic acid into the catalyst furthermore, and the polymer B of the molecular weight 18000 [about ] which has an average of 1.7 methyl dimethoxy silyl propyl groups per molecule at the end was compounded.

[0065][Example 6] Ethylene glycol was used as the initiator and the terminal hydroxyl groups of the polyoxypropylene diol produced by making the bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst react was changed into the allyloxy group. Methyl dimethoxysilane was made to react by having made chloroplatinic acid into the catalyst furthermore, and the polymer B of the molecular weight 18000 [about ] which has an average of 1.7 methyl dimethoxy silyl propyl groups per molecule at the end was compounded.

[0066][Example 7] Glycerin was used as the initiator and the terminal hydroxyl groups of the polyoxypropylene triol produced by making the bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst react was changed into the allyloxy group. Methyl dimethoxysilane was made to react by having made chloroplatinic acid into the catalyst furthermore, and the polymer C of the molecular weight 18000 [about ] which has an average of 1.8 methyl dimethoxy silyl propyl groups per molecule at the end was compounded.

[0067][Example 8] Glycerin was used as the initiator and the terminal hydroxyl groups of the polyoxypropylene triol produced by making the bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst react was changed into the allyloxy group. Methyl dimethoxysilane was made to react by having made chloroplatinic acid into the catalyst furthermore, and the polymer D of the molecular weight 18000 [about ] which has an average of 2.4 methyl dimethoxy silyl propyl groups per molecule at the end was compounded.

[0068][Example 9] Glycerin was used as the initiator and the terminal hydroxyl groups of the polyoxypropylene triol produced by making the bottom propylene oxide of existence of a zinc hexa cyanocobaltate complex compound catalyst react was changed into the allyloxy group. Methyl dimethoxysilane was made to react by having made chloroplatinic acid into the catalyst furthermore, and the polymer E of the molecular weight 30000 [about ] which has an average of 2.4 methyl dimethoxy silyl propyl groups per molecule at the end was compounded.

[0069][Examples 10-18] Polymer A-E and low molecule polymer a-d (or dioctyl phthalate: DOP) were mixed at a rate of the description to Table 1, mixed liquor was obtained, and the viscosity (a unit is cP) at 25 \*\* was measured. P-Si shows the number of the hydrolytic silicon groups per molecule of a polymer among front.

[0070]As opposed to these 160 copies of mixed liquor --- calcium carbonate (Shiroishi calcium company make.) 75 copies of Hakuanka GCR, 75 copies of calcium carbonate (Shiroishi calcium company make, HOWATON SB), 30 copies of titanium dioxides, and stabilizer (the mixture of an antioxidant, an ultraviolet ray absorbent, and light stabilizer.) the Ciba-Gaigy make, tinuvin B752 copy, and a photo-setting resin (the Toagosei Chemical Industry Co., Ltd. make.) It kneaded under the conditions which add ARONIKUSU M60205 copy, two copies of silane coupling agents (the Shin-Etsu Chemical Co., Ltd. make, \*\*KBM803), one copy of hydrogenation castor oil, and two copies of dibutyl tin bisacetylacetonate (the Nihon Kagaku Sangyo Co., Ltd. make, NASEMUSUZU), and moisture does not mix, and was considered as the uniform mixture.

[0071]Although it pierced with the JIS No. 3 dumbbell after creating the sheet about 2 mm thick and carrying out cure for seven days at 50 \*\* subsequently for seven days at 20 \*\* --- a 50% modulus ( $M_{50}$  unit/kg/cm<sup>2</sup>) and breaking strength (unit: kg/cm<sup>2</sup>) --- it was extended and (%) was measured.

[0072]After having applied the solvent system alkyl paint (the Rock Paint Co., Ltd. make, house paint) after creating and carrying out cure of the sheet about 1 cm thick, and heating for one week at 70 \*\*, it was exposed to the outdoors and the dirt situation of the paint surface was observed one

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2. \*\*\* shows the word which can not be translated.
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## CORRECTION OR AMENDMENT

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[F]

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[Written Amendment]

[Filing date] Heisei 13(2001) December 27 (2001.12.27)

[Amendment 1]

[Document to be Amended] Description

[Item(s) to be Amended] Claims

[Method of Amendment] Change

[Proposed Amendment]

[Claim(s)]

[Claim 1] As opposed to polymer with an or more 8000 molecular weight [ or less 50000 ] (I) and polymer (I) 100 weight section which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups, A room-temperature-curing nature constituent containing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II) 1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups.

[Claim 2] The room-temperature-curing nature constituent according to claim 1 whose both main chain of polymer (I) and main chain of low molecule polymer (II) are polyether intrinsically.

[Claim 3] The room-temperature-curing nature constituent according to claim 1 or 2 in which a room-temperature-curing nature constituent does not contain a plasticizer substantially.

[Claim 4] The room-temperature-curing nature constituent according to claim 1 or 2 which a room-temperature-curing nature constituent contains a plasticizer further, and does not contain a low molecule plasticizer as the plasticizer.

[Claim 5] As opposed to polymer (I) 100 with an or more 8000 molecular weight [ or less 50000 ]

weight section which averages in one molecule and has 1.5 or less one or more piece hydrolytic silicon groups, A manufacturing method of a room-temperature-curing nature constituent mixing less than the with an or more 300 molecular weight [ less than 8000 ] more than low molecule polymer (II) 1 weight-section 200 weight section that averages in one molecule and has 1.5 or less 0.5 or more piece hydrolytic silicon groups.

[Amendment 2]

[Document to be Amended] Description

[Item(s) to be Amended] 0002

[Method of Amendment] Change

[Proposed Amendment]

[0002]

[Description of the Prior Art] The method of making an end harden various kinds of polymers which have a hydrolytic silicon group, and using it for a sealing material, adhesives, etc. is known well, and is a useful method industrially.

[Amendment 3]

[Document to be Amended] Description

[Item(s) to be Amended] 0003

[Method of Amendment] Change

[Proposed Amendment]

[0003] The polymer especially whose main chain is polyether among such polymers is liquefied at a room temperature, and when a hardened material holds pliability also at low temperature comparatively and uses for a sealing material, adhesives, etc., it is provided with the desirable characteristic.

[Amendment 4]

[Document to be Amended] Description

[Item(s) to be Amended] 0007

[Method of Amendment] Change

[Proposed Amendment]

[0007] The hardenability constituent in which translatability adds a very low reactant plasticizer is proposed by JP 5-59267A, without reducing the pliability of a hardened material to the polymer of the hygroscopic-surface-moisture hardenability which has a hydrolytic silicon group in order to cancel such a fault. However, in the use to a sealing material etc., pliability is higher and it is the good characteristic of elongation, and when it paints on the surface, the hardenability constituent in which surface stain resistance has been improved is called for. Although various kinds of inorganic bulking agents are usually used in combination called a actual sealing material and adhesives, a polymer's own pliability is much more required for use of a bulking agent in order to harden the physical properties of a hardened material.

[Amendment 5]

[Document to be Amended] Description

[Item(s) to be Amended] 0008

[Method of Amendment] Change

[Proposed Amendment]

[0008]

[Problem(s) to be Solved by the Invention] Then, even if the viscosity of the hardenability constituent could be reduced and it used the inorganic bulking agent, the hardened material with a flexibly good extension characteristic was given, and moreover, translatability was dramatically low, and as a result of examining the additive agent which does not pollute a surface coat, it resulted in this invention.

[Amendment 6]

[Document to be Amended] Description

[Item(s) to be Amended] 0034

[Method of Amendment] Change

[Proposed Amendment]

[0034] Especially in manufacture of low molecule polymer (II), the molecular terminal group besides hydrolytic silicon Motomochi may be an inertness organic group. For example, it may manufacture by changing a terminal unsaturated group into an inertness organic group by the method of making terminal hydroxyl groups react to a hydrolytic silicon group with a benzoyl chloride etc. by using as a

raw material the polyether monoar which manufactured the terminal unsaturated group content mono- hydroxy compound as an initiator.

[Amendment 7]

[Document to be Amended]Description

[Item(s) to be Amended]0044

[Method of Amendment]Change

[Proposed Amendment]

[0044]The following compound can be used as a curing catalyst. Metal salt, such as an alkyl titanate, an organic silicon titanate, and bismuth tris-2-ethylhexanoate, Acidic compounds, such as phosphoric acid, p-toluenesulfonic acid, and phthalic acid, a butylamine, Aliphatic series monoamines, such as hexylamine, octylamine, decyl amine, and lauryl amine, Aliphatic diamine, such as ethylenediamine and a hexanediamine, diethylenetriamine, Aliphatic polyamine, such as triethylenetetramine and tetraethylenepentamine. Amine compounds, such as aromatic amine, such as heterocyclic amine, such as piperidine and a piperazine, and a meta-phenylenediamine, ethanolamines, triethylamine, and various denaturation amine used as a hardening agent of an epoxy resin.

[Amendment 8]

[Document to be Amended]Description

[Item(s) to be Amended]0051

[Method of Amendment]Change

[Proposed Amendment]

[0051]If it is considered as a bulking agent, the publicly known following bulking agent can be used. The calcium carbonate which carried out the surface treatment of the surface with fatty acid or a resin acid system organic matter. Colloid calcium carbonate with a mean particle diameter of 1 micrometer or less which furthermore impalpable-powder-ized this. Calcium carbonate, such as precipitated calcium carbonate with a mean particle diameter of 1-3 micrometers manufactured with the sedimentation method, and heavy calcium carbonate with a mean particle diameter of 1-20 micrometers, Fumed silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, carbon black, Magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, Powder state bulking agents, such as bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, a milt balloon, wood flour, pulp, a cotton chip, mica, blacking wash shell flour, chaff powder, graphite, aluminum impalpable powder, and the Flint powder. Fibrous fillers, such as glass fiber, a glass filament, carbon fiber, the Kevlar textiles, and a polyethylene fiber.

[Amendment 9]

[Document to be Amended]Description

[Item(s) to be Amended]0054

[Method of Amendment]Change

[Proposed Amendment]

[0054]As a plasticizer, for example Dioctyl phthalate, dibutyl phthalate, Phthalic acid alkyl ester, such as phthalic acid benzyl butyl ester, Dioctyl adipate, Aliphatic-carboxylic-acid alkyl ester, pentaerythritol ester, such as succinic acid diisodecyl, dibutyl sebacate, and butyl oleate etc.; Trioctyl phosphate, Phosphoric ester, such as tricresyl phosphate; epoxy plasticizer, chlorinated paraffin; such as epoxidized soybean oil and epoxy stearic acid benzyl, etc. can be independent, or can use it with two or more sorts of mixtures.

[Amendment 10]

[Document to be Amended]Description

[Item(s) to be Amended]0062

[Method of Amendment]Change

[Proposed Amendment]

[0062][Example 3] Propylene oxide was made to react by having used allyl alcohol as the initiator, and the polyoxypropylene monooar of the molecular weight 2000 was manufactured. Then, it was made to react to a hydroxyl group and the benzoyl chloride of equimolar under existence of triethylamine. The reaction mixture was diluted with 5-times the amount hexane, and was rinsed, except for the triethylamine hydrochloride, hexane was distilled off and one end acquired the monoallyl object of the benzoyloxy group. Next, it was made to react to methyl dimethoxysilane of the equivalent 80% to an allyl group by having made chloroplatinic acid into the catalyst, and the polymer c of the molecular weight 2200 which has a methyl dimethoxy silyl propyl group was

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compounded. Viscosity was 700cP at 25 \*\*.

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